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Some electronic instruments used in nuclear spectroscopic investigations

By BJÖRN ÅSTRÖM

With 17 figures in the text

SUMMARY

A description is given of two complete pulse-height spectrometers for electromagnetic radiations and alpha-particles respectively. The detectors and the electronic devices used for the counting and analyzing of the detector-pulses are described in some detail.

Introduction

In many nuclear investigations the amount of radioactive material available for measurements is very limited. The reason for this may be that the cross section for a certain nuclear reaction is low so that even a saturating bombardment in a cyclotron yields only a small number of atoms to be investigated. In other cases, where several isotopes of a certain element are formed in a cyclotron irradiation, it may be necessary to make an electromagnetic isotope separation before measurements. The yield of such a separation is rather low and the final samples are therefore weak. A third class of measurements where the counting rate is inevitably low, due to the nature of the measurement itself, is in coincidence experiments. Many experiments of the types described have been made in this laboratory. It was therefore necessary to build instruments suitable for the measurement of weak activities.

The purpose of the measurements has been to identify nuclides from their half-life and the energy of α-particles or γ-quanta emitted in their disintegration or to determine the values of these quantities in cases where they are not known [1]. Coincidence measurements and determination of the half-lives of excited states have also been made [2, 3].

If sufficiently strong sources are available, the energy of γ-quanta can be measured with a precision up to 1 part in 10^4 by means of magnetic β-ray spectrometers or bent-crystal gamma spectrometers [4]. The same precision can also be obtained in α-energy measurements using magnetic spectrometers [5]. However, the high precision in energy determination is obtained at the expense of the transmission in the spectrometer, because only radiation leaving the source within a very small solid angle is registered. In the present state of technique the upper limit of transmission in ordinary β-ray spectrometers seems to be about 10 % and the resolution at this transmission should then be of the order of 3 % [4]. It must also be pointed out that if one wants to correlate energy and time measurements, an electrical detector must generally be used in the above spectrometers. The spectrum is then measured one point at a time and one thus has a "single-channel" instrument, which only records a very small fraction of the total number of disintegrations in the sample. With semicircular spectrometers and photographic detection "multi-channel" measurements are made by recording a whole spectrum at the same time, but only very rough time determinations are possible by changing the photographic plates at intervals. The poor utilization of the sample still remains due to the low solid angle.

From the above it is obvious that other types of instruments must be used in nuclear spectroscopic measurements where only weak sources are available. For this type of measurements pulse-height spectroscopy has therefore been used.

Several types of radiation detectors such as ionisation chambers and scintillation counters deliver a voltage pulse, the amplitude of which is proportional to the energy of the particle or quantum being absorbed in the detector. The pulse-height distribution from such a detector can therefore be easily converted into an energy spectrum of the radiation.

The efficiency for absorption of α-particles in the gas of an ionisation chamber is 100 %. The efficiency of a NaI crystal in a scintillation detector is also almost 100 % for X-rays and low energy γ-rays and decreases at higher energies at a rate depending on the size of the crystal. The solid angle over which the detector sees the source can in both cases easily be made to approach 50 %, and with the scintillation detector it can be made even higher by placing the sample inside the crystal.

If the pulse-height distribution is measured with a multichannel analyzer, a pulse-height spectrometer will therefore record a fraction of the total number of events in the sample that is only determined by the solid angle, and, for high energy γ-rays, also by the decreasing crystal efficiency.

The energy resolution of a pulse-height spectrometer is lower than for the corresponding magnetic spectrometers. The width at half maximum of an α-peak from an ionisation chamber is of the order of 50 keV (Fig. 15) and the relative width of a 500 keV γ-peak is about 11 % (Fig. 2). In the last case the resolution varies inversely with the square root of the energy.

It is the purpose of this paper to describe some detectors and electronic instruments that have been used for pulse-height spectroscopic measurements. A more complete treatment of different parts of the subject has been given for instance by A. Ghiorso *et al.* [6], P. R. Bell [7], and J. B. Birks [8].

Experiments involving pulse-height spectroscopy

In a series of experiments the half-lives of isomeric states in lead and thallium isotopes have been measured [2, 3]. These measurements also included delayed-coincidence measurements. The selection of gamma energies was obtained by scintillation detectors and single-channel pulse-height analyzers. Both separated and unseparated sources were utilized and an automatic scintillation spectrometer was used for the determination of the relative isotopic abundances in the samples. The basic electronic equipment used in these measurements will be described.

The 225-cm cyclotron at the Nobel Institute has been used to accelerate heavy ions [9]. To study reaction mechanisms and eventually produce new isotopes these heavy ion beams (C^{12}, C^{13}, O^{16}, Ne^{20}, Ne^{22}) have been utilized for the irradiation of different targets of heavier elements (W, Au, U). In some cases the reaction products were separated from the irradiated targets by chemical methods [1] or by evaporation [10], and in other experiments a thin target was used and the recoiling reaction products were caught in a catcher foil behind the target [11].

The fusion of a light and a heavy nucleus often yields a neutron-deficient product nucleus, not only because of the ratio of the total number of neutrons to the total number of protons of the two reacting nuclei but also because the deexcitation of the compound-nucleus takes place by neutron evaporation. Many neutron-deficient isotopes among the heavy elements are α-radioactive. After the element number of a produced nuclide has been established by chemical methods, it is therefore often possible to determine its mass number from the α-energy and half-life. Even unknown mass numbers can be estimated in this way by means of the well established α-systematics [12]. The measurement of α-energies is therefore necessary in experiments of this type. The amount of activity produced is often very low due to low cross sections, and the measurements are being made by means of an ionisation chamber and a 50-channel pulse-height analyzer, which will be described below. The results of these experiments, which have been teamworks, will be published elsewhere.

Scintillation detectors for γ- and X-rays

A standard arrangement of a scintillation detector for spectroscopic measurements is shown in Fig. 1 together with the circuit of the preamplifier attached to it.

Several types of photomultipliers such as EMI 6260, Du Mont 6292 and RCA 6342 have been used with about the same results as far as energy resolution is concerned. With unselected tubes a relative full-width at half maximum of about 9 % was obtained for the 661 keV peak from a Cs^{137} sample. The RCA tube was preferred in coincidence arrangements [3] because of its lower spread in transit time. The potential distribution between the cathode, focusing electrode and the first dynode, giving the best resolution, was determined experimentally for each individual tube. The resistor values at the lower end of the voltage divider given in Fig. 1 could therefore vary slightly.

All NaI (Tl) crystals used have been of the aluminum-canned type, manufactured by Harshaw Chemical Co. Below are listed four different crystals together with a short description of their special features for certain types of measurements.

1. ∅ $1\frac{1}{2}'' \times \frac{1}{2}''$.—This crystal has the lowest background and is best suited for X-ray measurements Up to 100 keV photon energy the efficiency is almost 100 % but decreases rapidly at higher energies.

2. ∅ $1\frac{1}{2}'' \times 1''$.—This is a general purpose crystal for which the efficiency has been calculated [7]. Experimentally it was found to drop from 100 % at 100 keV to 10 % at 700 keV. The method used for determination of crystal efficiencies has been described in [13].

3. ∅ $1\frac{3}{4}'' \times 2''$.—This is the biggest crystal which can be used with a 2″ photomultiplier. It is mainly used when one is interested in high-energy gammas. The Na^{22} spectrum measured with this crystal is shown in Fig. 2A.

4. ∅ $1\frac{3}{4}'' \times 2''$ hollow crystal.—A high solid angle is obtained which favours the appearance of sum-peaks of coincident gamma-rays. This can give some information about the energy levels in decay schemes. The coincidence relations in the Na^{22} spectrum are demonstrated in Fig. 2B. This crystal has also been used for delayed coincidence measurements [2].

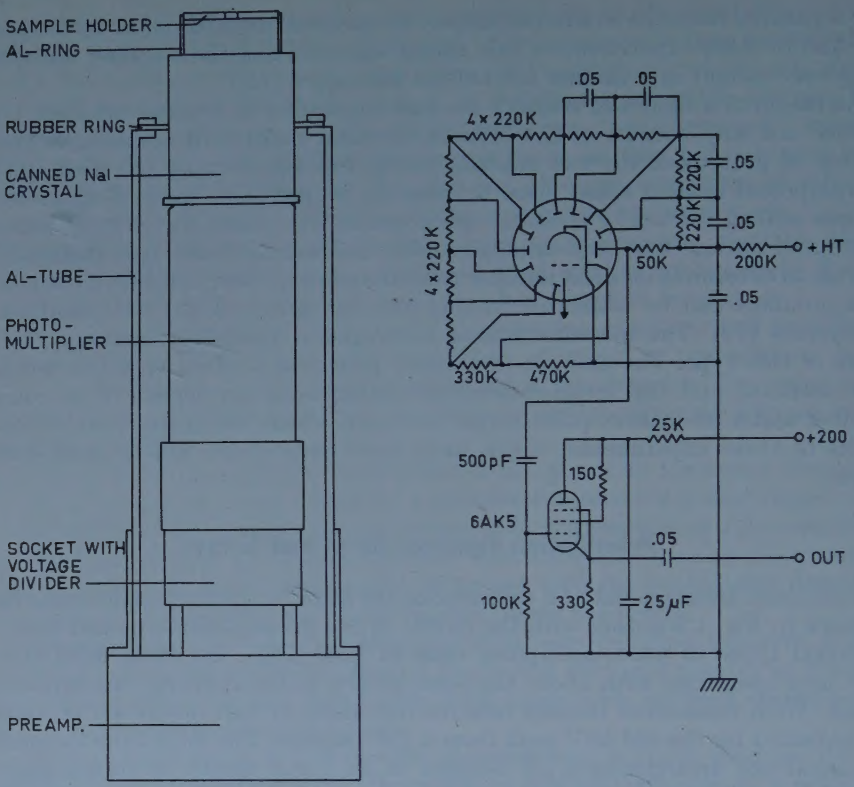

Fig. 1. Scintillation detector and circuit of preamplifier. Mechanical layout and circuit vary slightly for different types of photomultipliers, see text.

In some cases it was found that the pulse size was not linearly proportional to the γ-energy if the collector pulses were bigger than 0.25 volts. The high tension was therefore so chosen that pulses to be measured did not exceed this size. Since the range of the pulse-height analyzer was 100 volts, the gain of the linear amplifier between the detector and the analyzer was necessarily greater than 400 times.

Table 1. Background of scintillation detectors. Counts/min in a 2 % channel, set at 100 keV and 1 MeV respectively.

Crystal	γ-energy keV	Shielded	Unshielded
1½″ × ½″	100 1000	7 3	100 5
1½″ × 1″	100 1000	10 5	200 10
1¾″ × 2″	100 1000	30 15	400 50

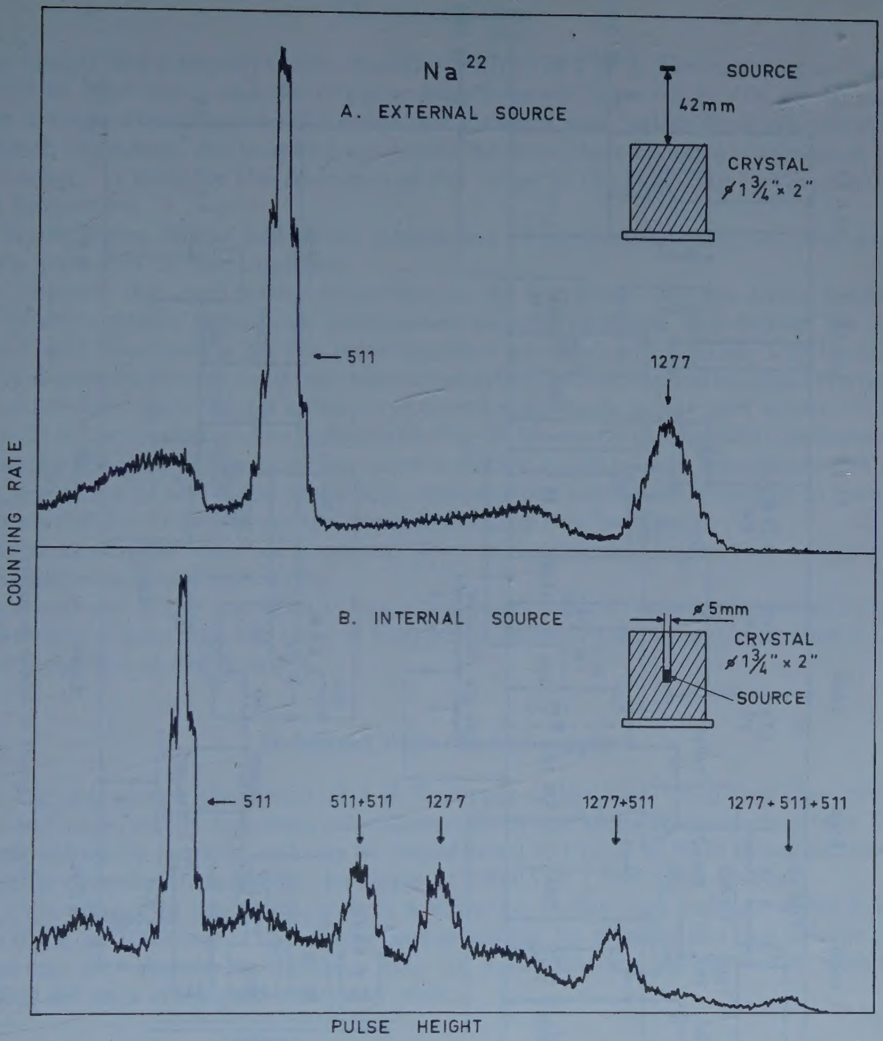

Fig. 2. Scintillation spectra, measured with a Na^{22} source. The channel width of the pulse-height analyzer was 2 % and the amplifier gain was changed between the two runs. Energies indicated are in keV.

In order to lower the background when weak samples are measured, a graded shield is used. This shield consists of 15 mm of lead, 1 mm of cadmium and 1 mm of copper. Table 1 gives the background of the different crystals measured in an ordinary laboratory room.

One method of mounting samples is indicated in Fig. 1. The active material is placed in a 5-mm hole in a perspex disk, resting on an aluminum ring. This gives a well reproducible geometry with low back scattering.

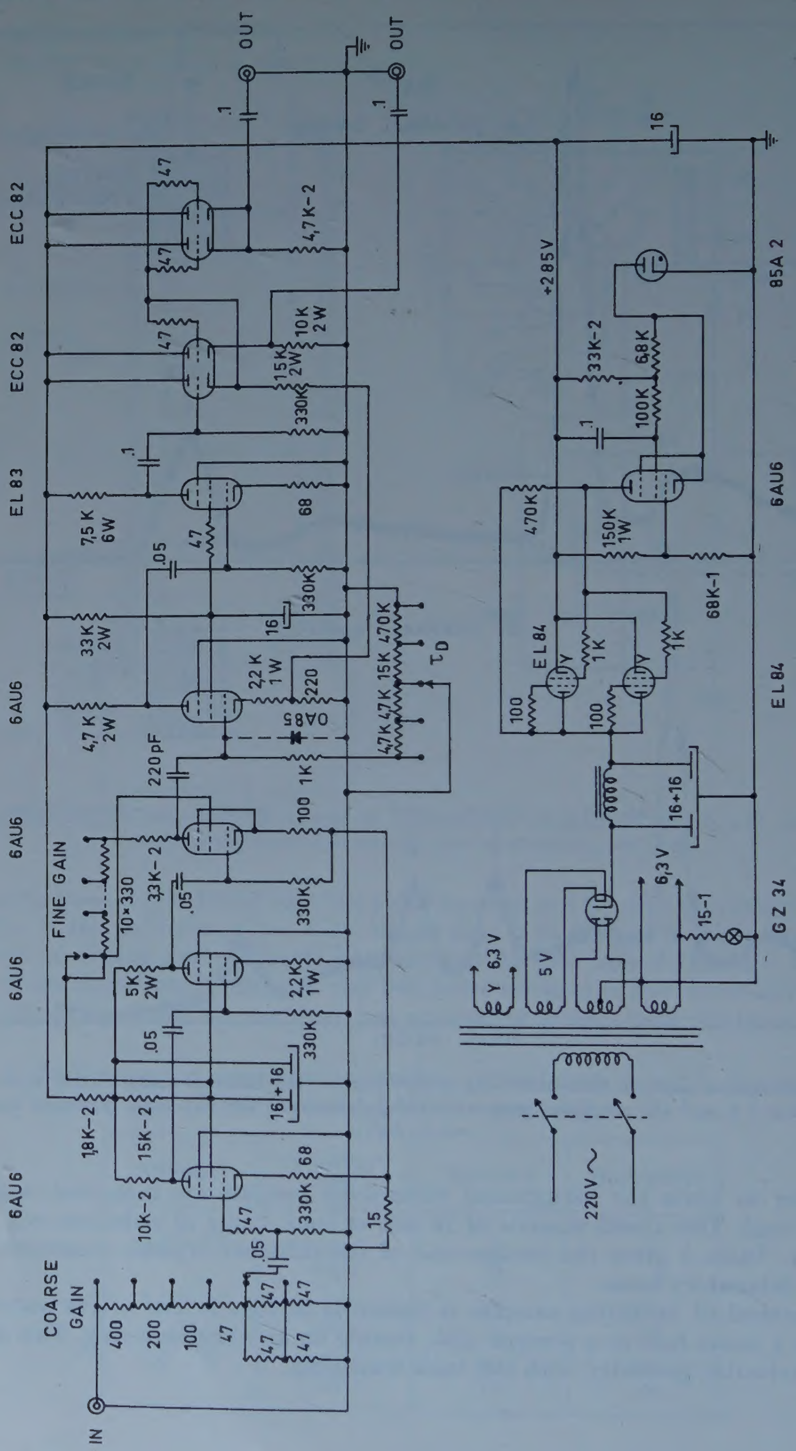

Fig. 3. Circuit of linear amplifier.

Linear amplifier

The circuit of a standard linear amplifier is given in Fig. 3. The gain can be varied from 50 to 2500 times and the clipping time constant from 0.3 to 100 μs. In cases where a longer time constant is needed, for instance with pulses from ion-collecting ionisation chambers, the coupling condenser between the two loops is increased and a d.c. supply is used for the filaments of the tubes in the first loop to keep the 50-cycle hum down.

In applications where additional integration of pulses is needed, the integrating filter is placed in the preamplifier.

To improve the overloading properties of the amplifier, the two tubes working with positive pulses have large unbypassed cathode-resistors. This lowers the grid current and thus speeds up the recovery after an overloading pulse. The blocking time is determined by the clipping time constant (τ_D). The overloading characteristics can be further improved by adding a d.c.-restoring diode in the grid circuit of the first tube in the second loop as indicated in Fig. 3. However, this has the disadvantage of making the gain of the amplifier more sensitive to changes in temperature as the back resistance of the diode has a high temperature coefficient. Changes in gain at this point of the amplifier are not compensated by any feed-back.

The peak-to-peak value of noise and 50-cycle ripple at full band width and gain of the amplifier is about 0.25 volts.

The response of the amplifier is linear for pulses with an amplitude at the output of 100 volts or less. The rise time of the output pulse (10–90 %) is 0.3 μs when a fast pulse is applied at the input.

Stabilized high tension supply

In Fig. 4 is shown the circuit of a H.T. supply of the high frequency type, which has been used for GM counters, scintillation detectors and ionisation chambers. The output voltage is positive and can be varied from 400 volts to 2500 volts. Maximum available current is 1 mA, and the ripple at 1000 V is 1 volt peak-to-peak.

A 10 % change in line voltage gives a variation in the high voltage which is less than 0.1 % at 1000 volts. This figure was measured by varying the line voltage and observing the change in counting rate in a scintillation spectrometer with the channel set on a steep part of a γ-ray peak.

Scalers

At moderate counting rates the dekatron scaler of Fig. 5 is used to count pulses. It has a time resolution of 200 μs and the input sensitivity can be varied between 0.2 volts and 30 volts. If high voltage is supplied into the connector marked HT, a self-quenching GM tube can be connected directly at the input. The octal connector shown at the input makes it possible to attach a preamplifier and/or a timer. In cases where higher counting rates must be handled, the fast prescaler of Fig. 6 is inserted before the dekatron scaler.

This scaler was designed so that the mechanical register could be run close to maximum speed (20 counts/sec) without the loss of pulses exceeding 1 % in any part of the system. Following the theory of Alaoglu and Smith [14] it was found that a

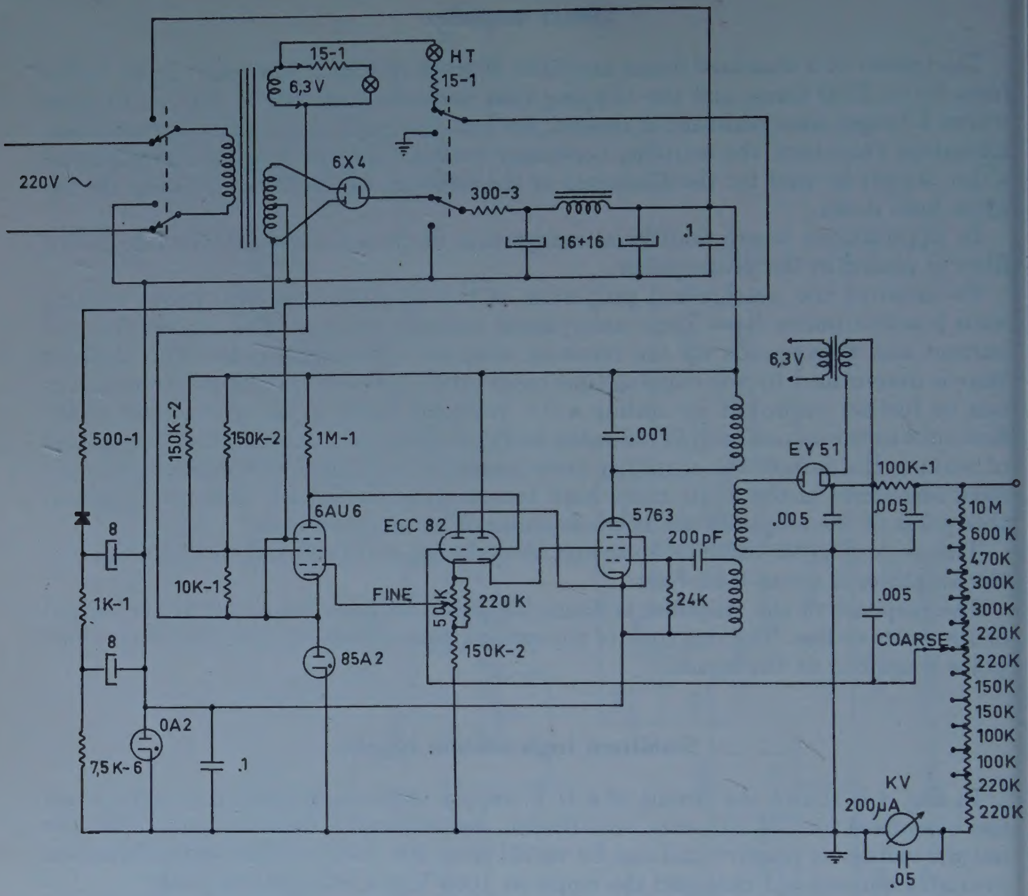

Fig. 4. Circuit of stabilized H.T. supply. Special connections at switches make output voltage zero in off position.

scale-of-8 with 1 μs resolving time gives optimum conditions. For 1 % loss the maximum counting rate at the input is 10000 counts/sec and the register is then running at 12.5 counts/sec. For the dekatron scaler alone the corresponding figures are 50 counts/sec and 0.5 counts/sec.

Electronic timer

The dekatron timer showed in Fig. 7 has been used to control scalers, generate time-markers in decay measurements and control the automatic scintillation spectrometer described in next section. The power line frequency is scaled down and used to close and open a mechanical relay at time intervals ranging from 2–2000 seconds. The timing accuracy is thus dependent of the accuracy of the line frequency. For all

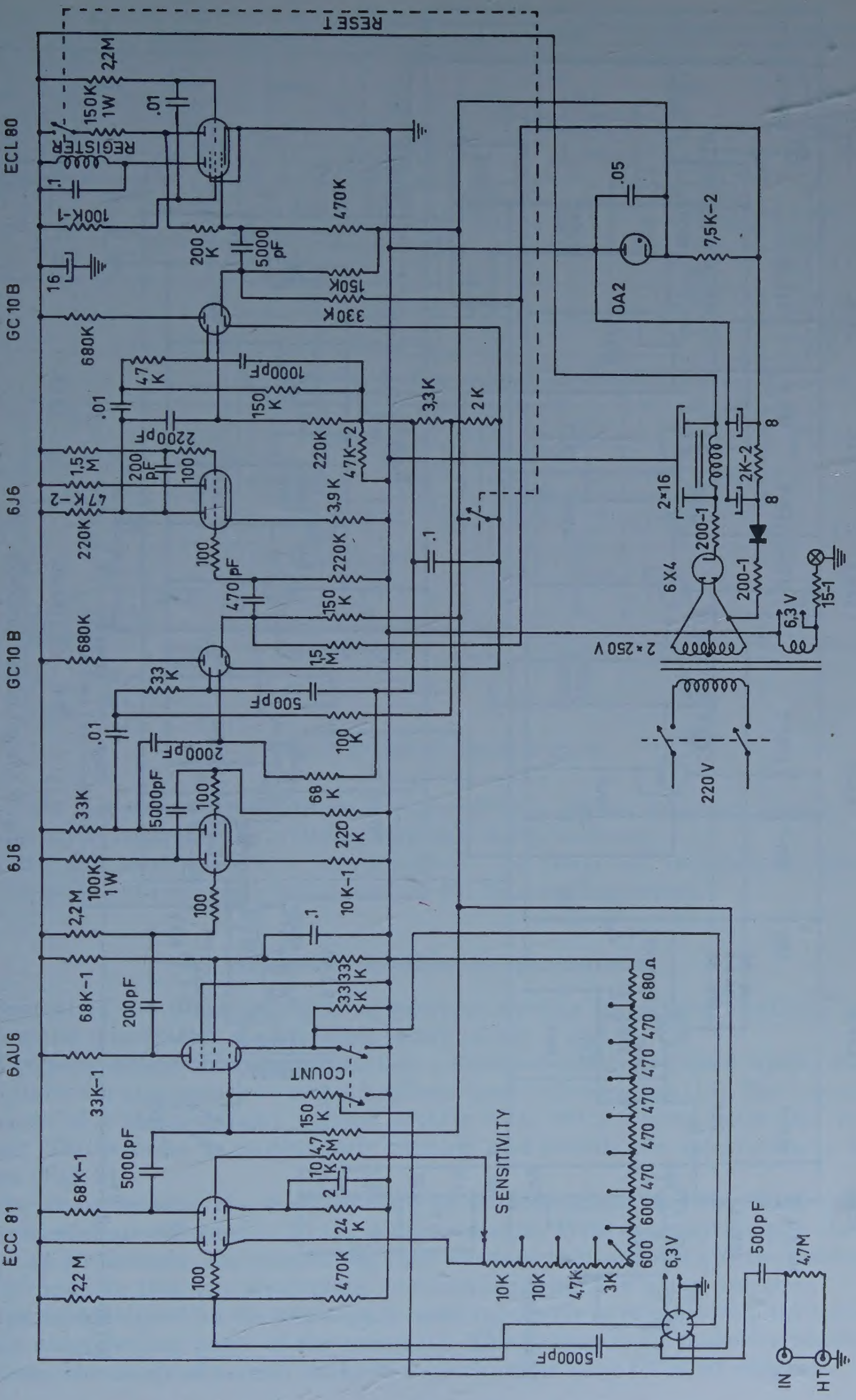

Fig. 5. Circuit of dekatron scaler.

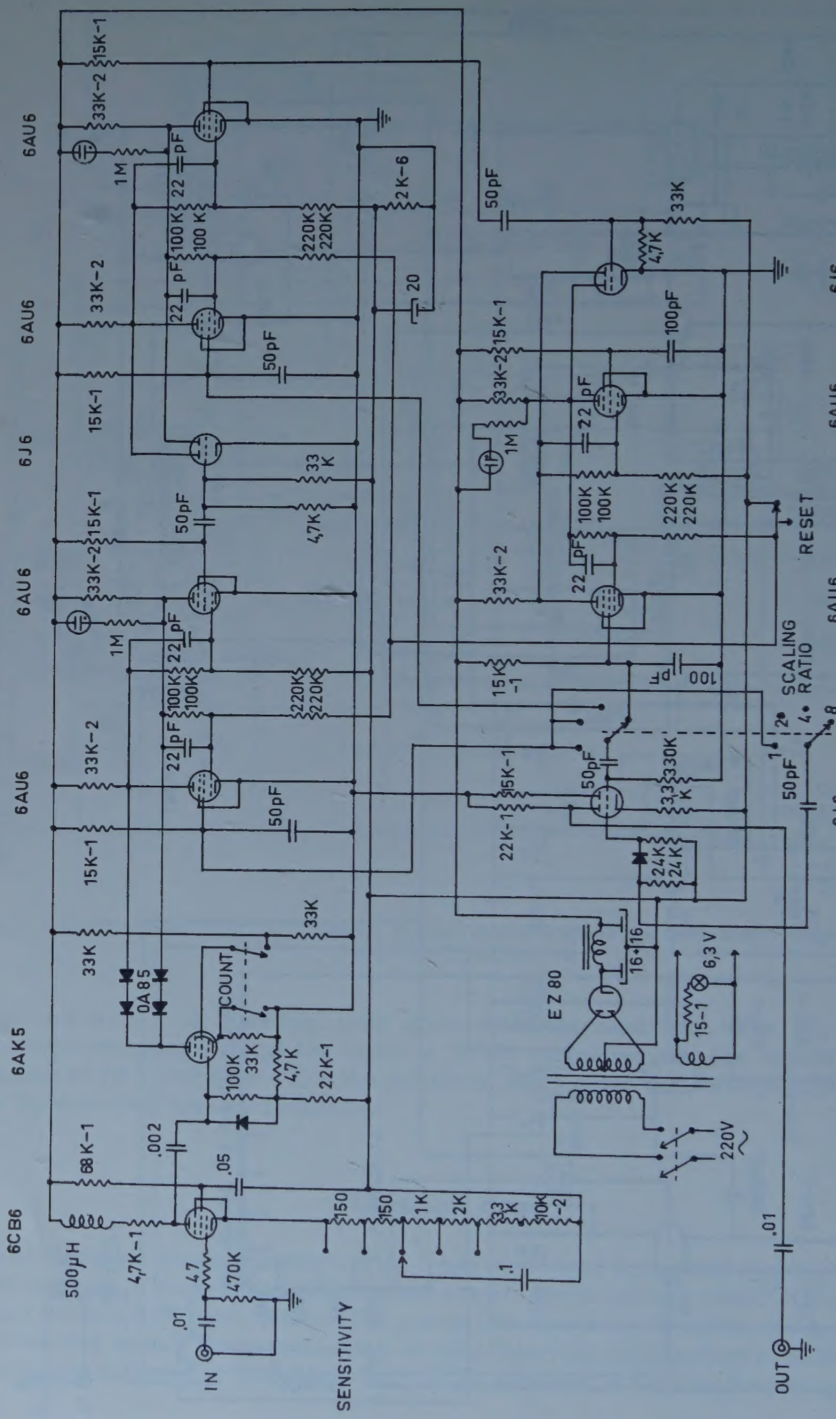

Fig. 6. Circuit of fast prescaler.

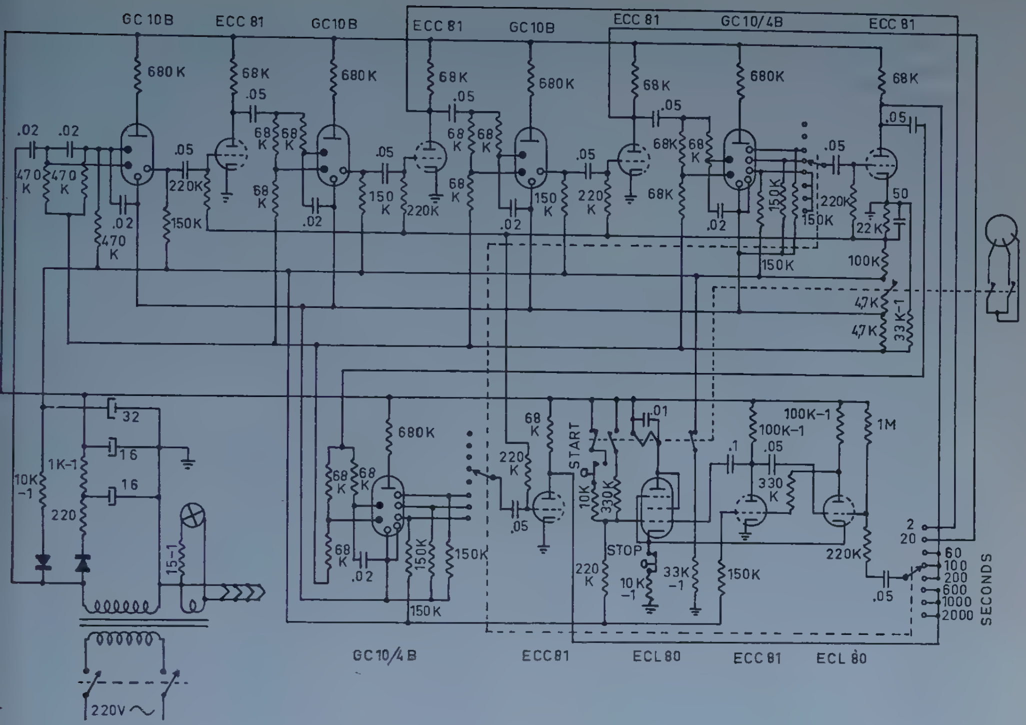

Fig. 7. Circuit of electronic timer.

accurate measurements the timer is powered by a special generator system whose frequency is controlled by a tuning fork with 0.1 % accuracy.

With the starting button shortcircuited, the timer will recycle automatically, giving pulses at intervals, determined by the time-setting switch.

Automatic scintillation spectrometer

Several of the units described in previous sections have been combined in the automatic scintillation spectrometer shown in Fig. 8 and 9.

The pulse-height analyzer which has a constant relative channel width, making it suitable for automatic recording, has been described elsewhere [15]. The bias supply consists of a stepping-relay voltage divider with 100 positions, controlled by the timer. The recorder is continuously running and records the spectrum as a histogram (Fig. 2).

The timer setting, the time constant of the rate meter and the chart speed of the recorder are adjusted to fit the source strength. With ordinary samples the time is set at 40 seconds, the integrating time constant is 10 seconds and the chart speed is 12″/hour. In this way a complete spectrum is recorded in about one hour.

Special "jumpers" in the bias supply make it possible to exclude from the measurement uninteresting parts of the spectrum. This arrangement makes it possible to measure the decay of certain peaks in a spectrum at more frequent intervals.

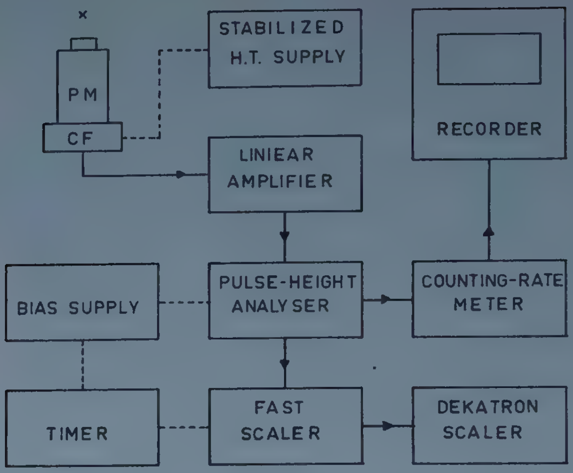

Fig. 8. Block diagram of automatic scintillation spectrometer. A Brown Potentiometer Recorder and an Ekco Counting Rate Meter, type 1037 A are used. Other units are described in text.

The scalers are used for manual measurements and when adjusting the spectrometer for an automatic run.

To ensure a good operational stability with small shifts in the calibration, the temperature of the spectrometer room is controlled to within ± 1°C and the mains supply is stabilized to ± 0.1 %.

For certain measurements requiring high stability over long periods of time, the stabilizing arrangement described by de Waard [16] is used.

The spectrometer is calibrated with standard sources and some isotopes which have proved to be useful for this purpose are listed in Table 2.

Table 2. Calibration sources for scintillation spectrometer.

Isotope	Photon energy, keV
Co^{60}	1332, 1172
Na^{22}	1277, 511
Zn^{65}	1120
Bi^{207}	1064, 569, 74
Mn^{54}	840
Cs^{137}	661, 32

Ionisation chamber for α-particles

The ionisation chamber shown in Fig. 10 has been used to analyze α-radioactive isotopes produced in heavy-ion cyclotron irradiations. The samples are ordinarily prepared on 25 mm platinum disks which fit into the positions on the sample holder. Five positions are available which means that the chamber can be loaded with five different samples which can be measured one at a time by turning the sample holder.

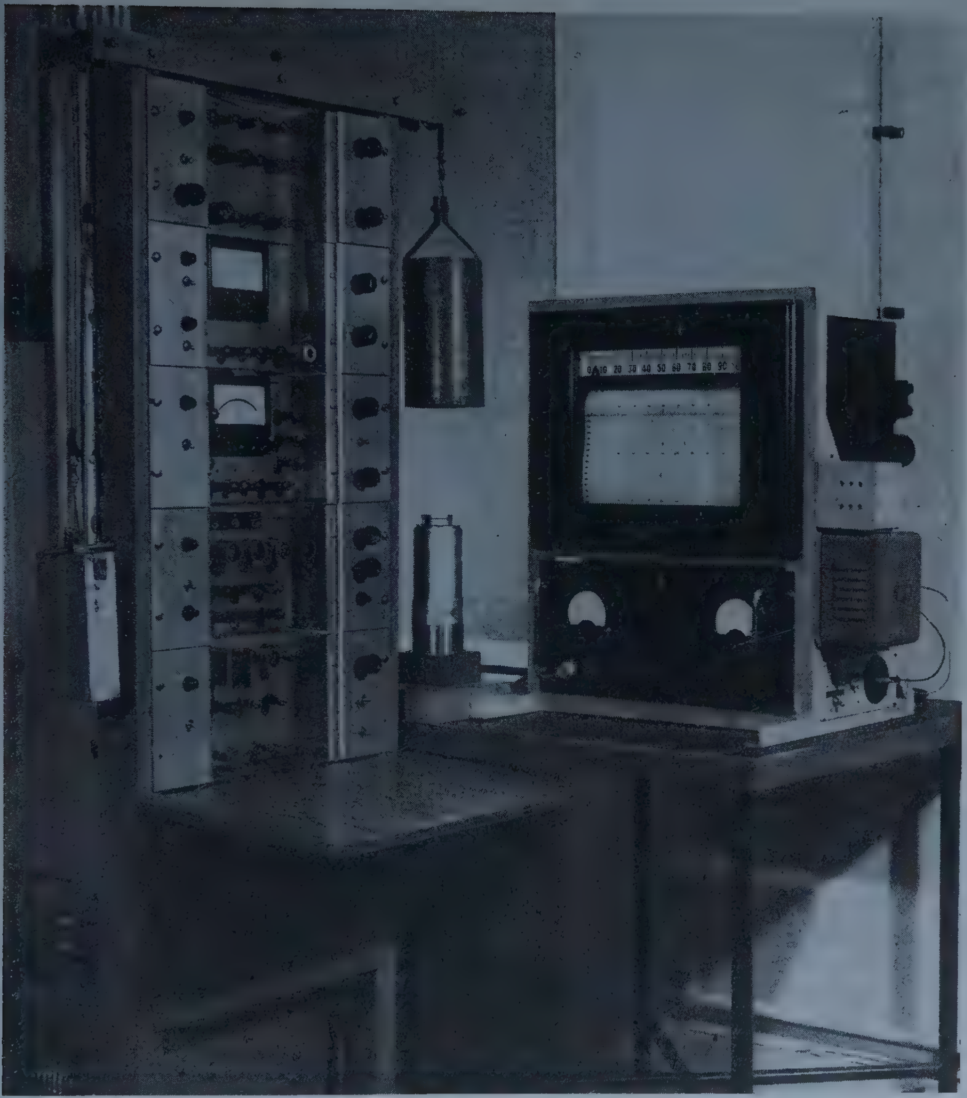

Fig. 9. Automatic Scintillation Spectrometer. The detector, with the lead shield lifted, is seen between the recorder and the rack with the rest of the electronic equipment.

One of the positions is generally used for a standard sample. The source electrode and the grid are supported by Teflon rods from the top of the chamber.

Samples are introduced by loosening four screws fixing the bottom plate to the wall and lifting the upper part of the chamber. The sample holder is then accessible. The grid is made of 0.1 mm steel wires spaced 1 mm and soldered to a brass ring. Different shapes of collector electrode have been tried but the small disk shown in the figure has given the best energy resolution. A guard electrode inserted between

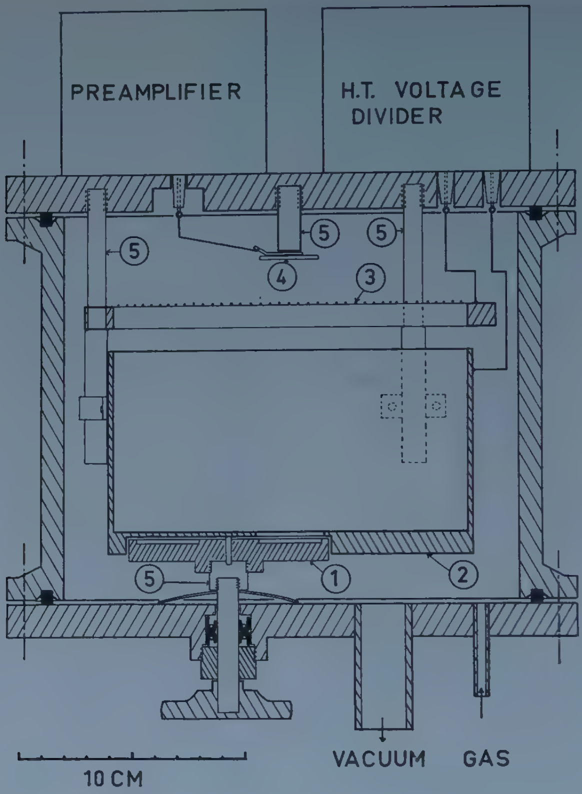

Fig. 10. Ionisation chamber. (1) Five-position sample holder. (2) Source electrode. (3) Grid. (4) Electron collector. (5) Teflon insulators.

the collector and the chamber wall and connected to the cathode of the first tube in the preamplifier increased the pulse amplitude, but did not improve the signal to noise ratio.

The chamber is evacuated by a mechanical pump through a short tube and a valve with a wide opening.

Preamplifier for ionisation chamber

With the present technique the ultimate limit of energy resolution is determined by amplifier noise. Much effort has therefore been spent on designing a low-noise amplifier system following principles given by Engelkeimer and Magnusson [17] and Gillespie [18].

The circuit given in Fig. 11 shows the preamplifier used together with the ionisation chamber just described.

The input 6AK5 tube was selected for low noise from about 25 tubes. With this particular tube a floating grid was found to be best. The integrating and differentiating time constants (in grid circuit of output cathode-follower and between loops in main amplifier, respectively) were then adjusted in combination with different values

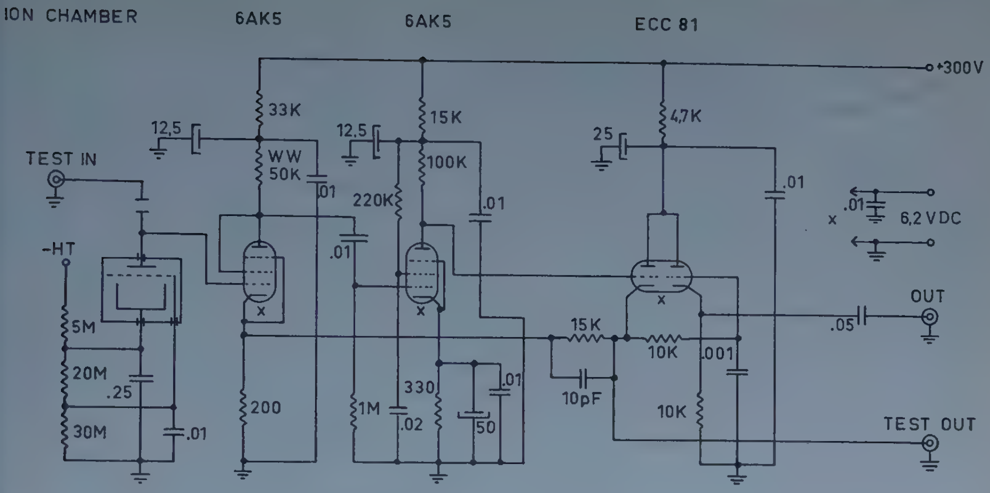

Fig. 11. Circuit of isonisation chamber and preamplifier.

of gas pressure and potential distribution between source electrode, grid and collector in the chamber. The highest possible total voltage which did not give spurious counts was used. In this way the optimal conditions were finally found. These were:

Gas pressure	3 atm.	Integrating time constant	10 μs
Source electrode potential	2800 volts	Differentiating time constant	10 μs
Grid potential	1700 volts		

To distinguish between peak broadening due to chamber and source effects, and amplifier noise respectively, pulses from a pulse generator were fed to the "test in" terminal. The "test out" terminal is used to inspect pulses before integration.

50-Channel pulse-height analyzer

The multichannel pulse-analyzer used for the measurement of α-energies is of the stacked discriminator type, based on the "snapper circuit" [19]. Each channel contains three tubes (6BN6, ECC 82, ECL 80) and a mechanical register. To trigger a channel it is necessary to supply the pulse to be measured and an inspection pulse. The bias of the channels increases sucessively by a potential difference corresponding to the channel width. Anti-coincidence circuits are so arranged that the pulse is only registered in the highest channel being triggered. A similar circuit is used in the stabilizer, Fig. 13. The maximum pulse height available for analysis is 250 volts and the channels can be set to cover one of the following amplitude ranges: 0–250, 0–125, 125–250, 0–50, 50–100, 100–150, 150–200 and 200–250 volts. This means that either a whole spectrum can be measured at a time or a smaller part of the spectrum can be investigated in greater detail.

A block diagram of the complete α-spectrometer is given in Fig. 12. The linear amplifier and preamplifier have already been described.

The pulse shaper consists of a condenser being charged by the incoming pulse via a diode and a thyratron discharging the condenser after the pulse has been registered

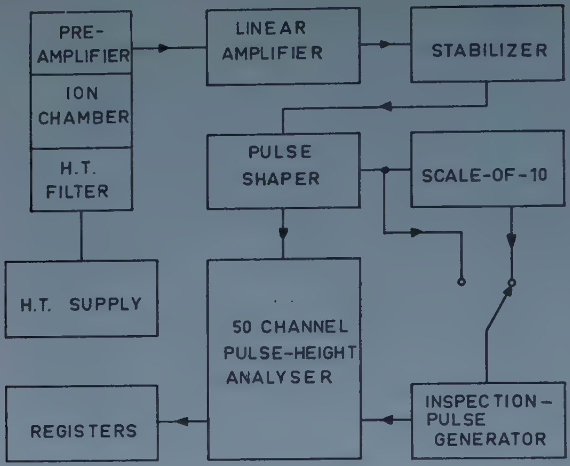

Fig. 12. Block diagram of pulse-height α-spectrometer.

in the pulse-height analyzer. The pulse so generated has a flat top and a length of about 40 μs. This time determines the time resolution of the sorting mechanism of the analyzer.

The channels in the analyzer are triggered by a 1 μs inspection pulse arriving from the inspection pulse generator 10 μs after the main pulse. The use of an inspection pulse is essential for the functioning of the snapper circuit and it has also other advantages. In coincidence experiments the inspection-pulse generator can be triggered by a second detector thus giving directly a coincidence spectrum. The maximum counting rate which the analyzer can handle is determined by the resolving power of the mechanical registers. The insertion of a scaler in the inspection pulse path as indicated in Fig. 12 has essentially the same effect as a corresponding scaler in each channel. The randomness in pulse distribution is smoothed out and the pulse-handling capacity in each channel becomes much higher.

The stabilizer (Fig. 13) has been incorporated to improve the over-all stability of the spectrometer when long runs are made. It consists of a motordriven attenuator controlled by the counting rates in two channels placed on different flanks of a reference peak in the α-spectrum. It keeps these two counting rates equal and thus locks all α-peaks to certain channels, independent of what causes a drift of pulse amplitude in the system. The pulses in the stabilizer channels are counted by dekatron tubes and when 10 or 100 pulses are accumulated in one channel the counting in that channel stops and the motor starts to change the amplification towards a higher counting rate in the other channel. When this channel has received the same number of pulses the motor stops and the cycle starts again. To avoid broadening of the peaks due to statistical fluctuations in counting rate and oscillations in the stabilizing characteristic, the time constants and damping of the system have been chosen so that several cycles of operation are needed for the compensation of a sudden change in amplifier gain.

The reference line for the stabilizer is obtained from a Po^{208} source ($E_\alpha = 5.10$ MeV) where the energy of the α-particles has been decreased below 5 MeV by absorption in an aluminum foil. To get a sharp peak even with the absorber, a hard collimation

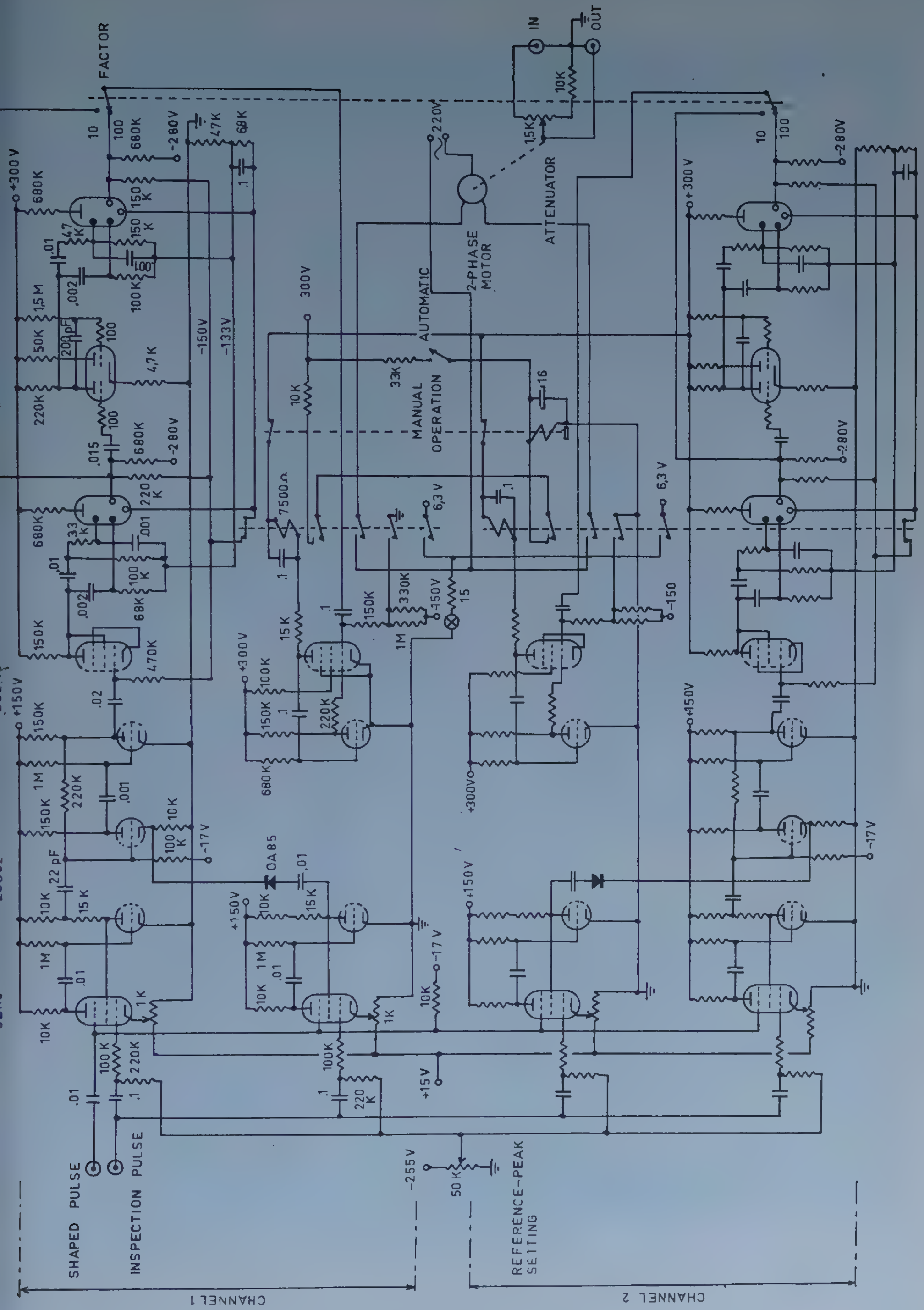

Fig. 13. Circuit of stabilizer used in α-spectrometer.

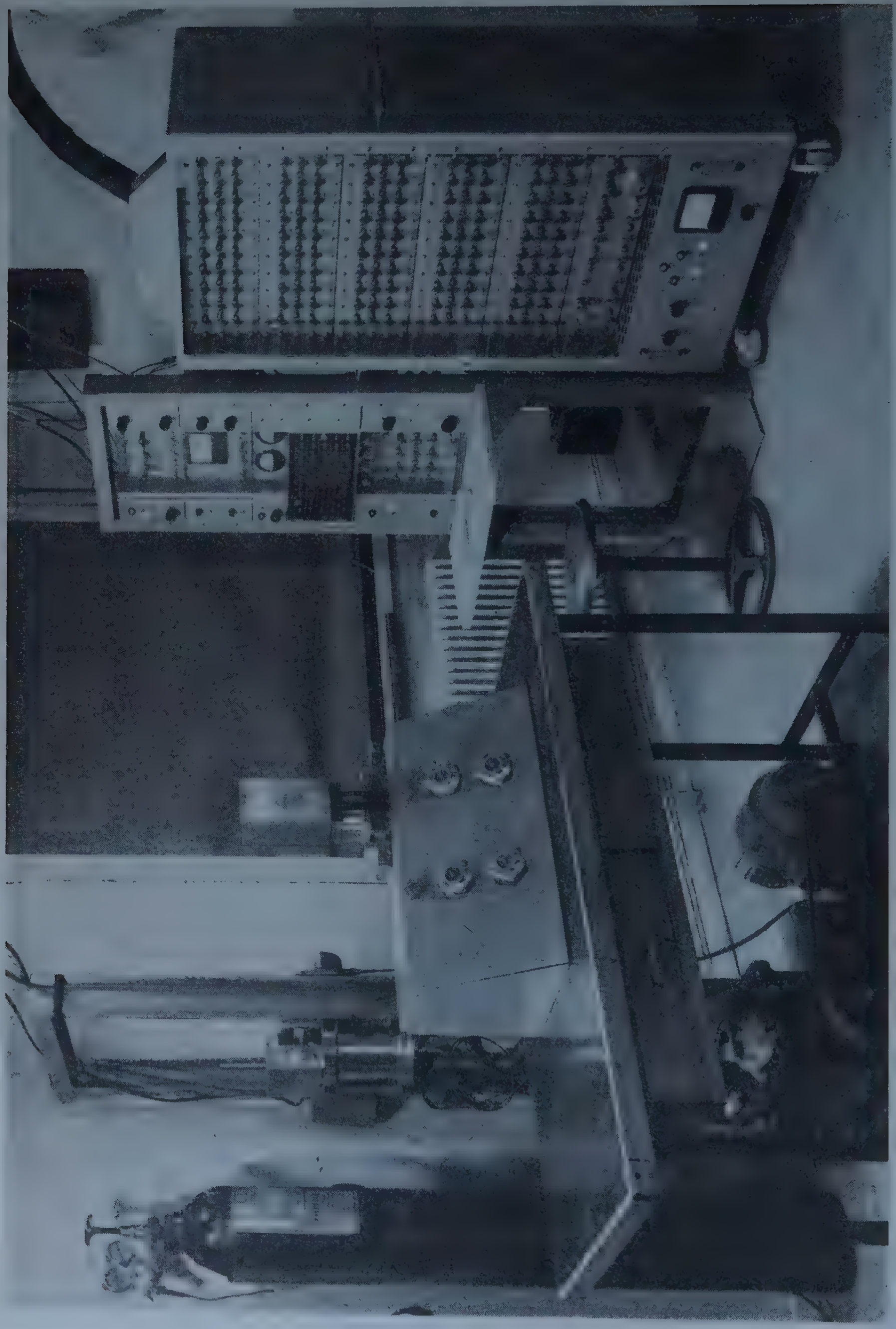

Fig. 14. Apparatus for the measurement of α-energies by pulse-height analysis. The ionisation chamber is mounted in the left rack and the 50-channel pulse-height analyzer is seen at the right. The second rack contains linear amplifier, H. T. supply, mechanical registers and stabiliser. (The small chamber on the table is not used in this work.)

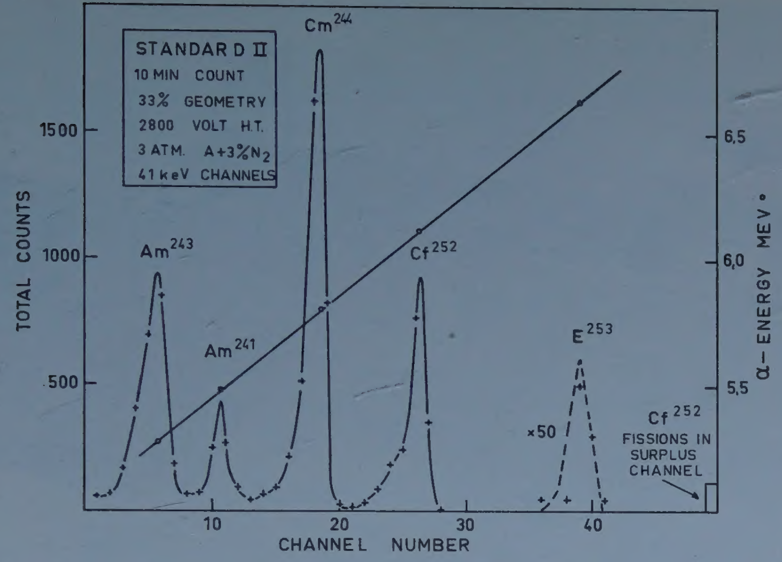

Fig. 15. The spectrum of a standard sample, used for the energy-calibration of the α-spectrometer.

is made by means of a Lektromesh screen [17], placed after the absorber. The reason for using this type of reference sample is that one wants an α-peak with a fairly high counting rate (about 200 cpm) but with an energy lower than any energy that one might want to measure. Still, some counts from the reference source are obtained in higher channels because of pile-up effects. Therefore, in measurements where one wants the lowest possible background, this source must be taken out of the chamber and the stabilizer can not be used.

Operation of the α-spectrometer

The gas used in the ion chamber is technical argon (99.9 % A, 0.003 % O_2) mixed with 3 % nitrogen [20]. The gas is taken directly from the cylinder without any additional purification. After a refilling, the energy resolution improves during the first hour and reaches a value corresponding to 55 keV full width at half maximum peak-height (Cm^{244} in Fig. 15). This resolution is then constant for several days of continuous operation. The reason for the change during the first hour is not known.

In most of the measurements made with the α-spectrometer a low background was essential. This was accomplished by avoiding the introduction of any activities belonging to the natural disintegration series into the chamber and by keeping to a minimum the time during which the interior of the chamber was exposed to air. After an accidental contamination, nickel plating of all parts reestablished the original conditions. The following numbers of counts at different energies were found during 24 hours, in a typical background measurement: 14 (6–7 MeV), 7 (7–8 MeV), and 1 (above 8 MeV).

The energy calibration of the spectrometer is made by means of standards. Fig. 15 shows the spectrum of one standard used. Other standards are Cm^{242} and Po^{210}.

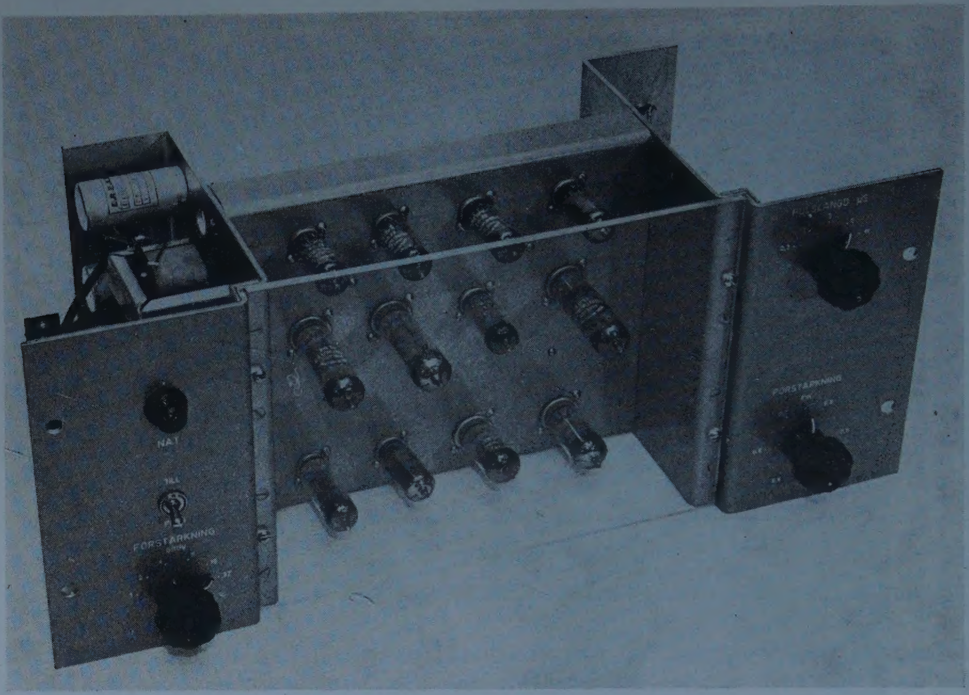

Fig. 16. The mechanical construction of a typical unit of electronic equipment (linear amplifier).

In order to get a high-energy calibration peak, a ThB source, giving the 8.78 MeV α-particles of ThC′, has been used on rare occasions. Great care was taken, not to contaminate the chamber with radio-thorium from which the source was prepared by the well known electro-deposition method.

The solid angle of the chamber was determined by measuring a sample that had previously been measured at Harwell, and was found to be 33 %. In some measurements where a lower solid angle could be tolerated, a little improvement in energy resolution was obtained by covering the sample with a disk, 2 mm thick and with a collimating hole, 8 mm in diameter.

Many samples measured with the spectrometer had a rather short half-life and it was necessary to start measurements as fast as possible. After some excercise it was possible to put the sample in the holder, close and evacuate the chamber, flush it twice, fill with gas and start counting in a little more than one minute.

Some remarks on the design and construction of electronic instruments

The instruments described in this paper have been used in various combinations in a great many investigations. Because of the variety of combinations necessary for different types of measurements, it has proved valuable to build the different devices as complete units with their own power supplies etc.

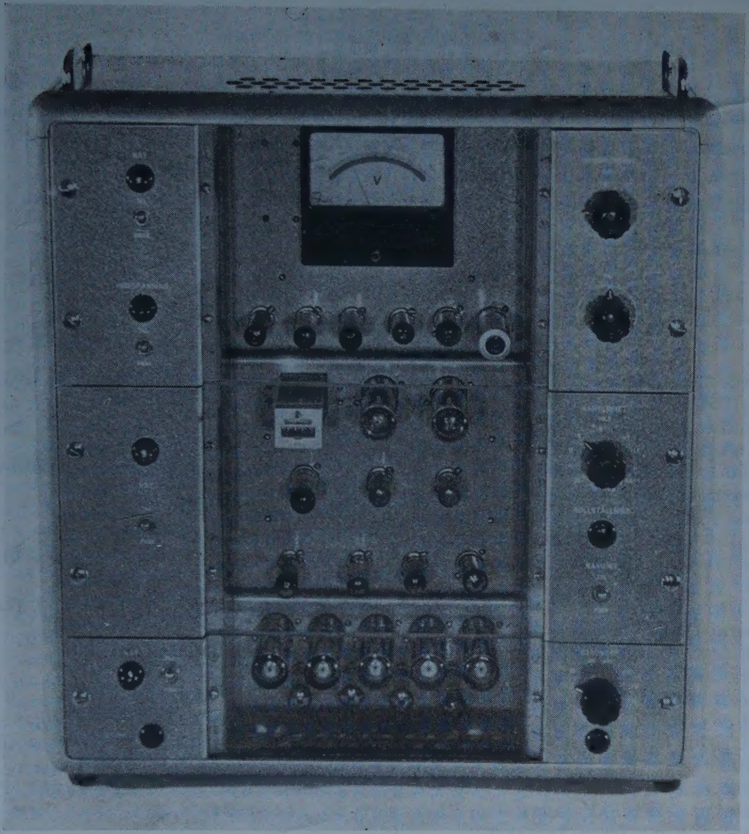

Fig. 17. H.T. supply, scaler and timer, used for GM-counting.

In order to facilitate maintenance and testing and to improve cooling, a special mechanical lay-out has been used in the instruments described. As can be seen from the photograph, Fig. 16, all tubes are mounted horizontally and are concentrated in a closed space where they can not heat other components. When several units are mounted on top of each other the tube compartments form a chimney, which is ventilated by natural circulation or, as in the 50-channel analyzer, by a blower. The power supply is mounted behind the left panel and switches and other controls are generally concentrated on the right panel. The rest of the components are easily accessible from the back where also all cable-connectors are placed. In bigger assemblies the units are placed in racks as in the automatic scintillation spectrometer (Fig. 9). For smaller assemblies, such as the scaler, high tension supply and timer used for GM counting (Fig. 17), an aluminum case is used.

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Nobel Institute for Physics, Stockholm 50.

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